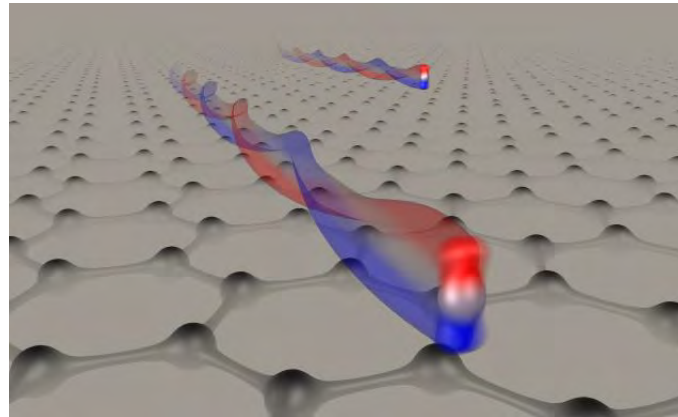


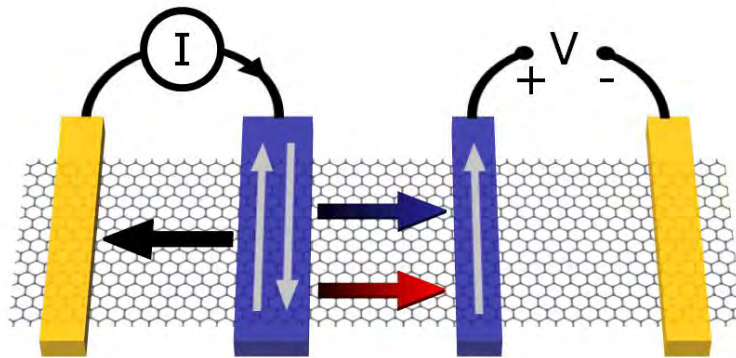
Novel architectures for graphene spintronics



Ivan Jesus Vera-Marun

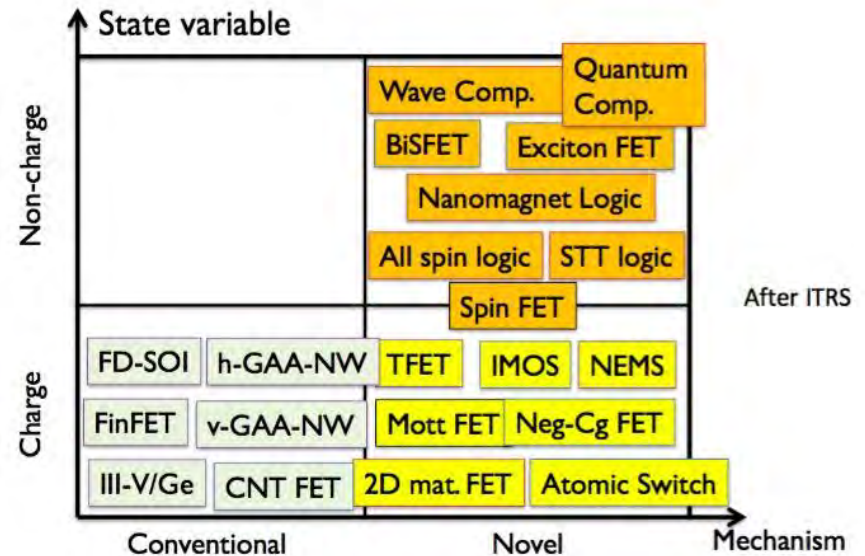
Nonlocal spin geometry

Novel spin functionality in future electronics (ITRS)
 Separate charge from other degrees of freedom
 All-electrical approach



Local charge current

Nonlocal pure spin currents, valley, heat...



What can we learn?

Double-gated encapsulated bilayer

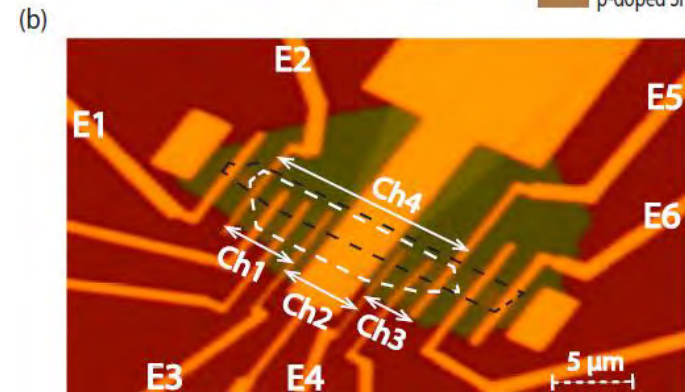
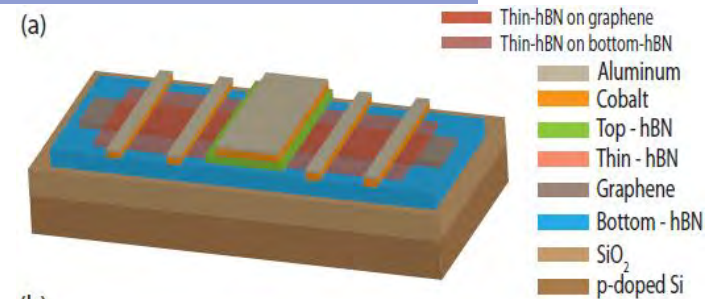
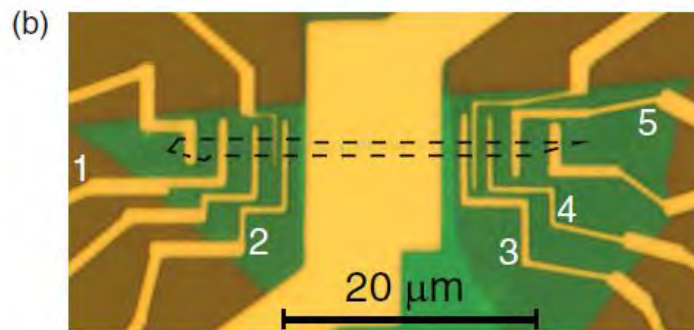
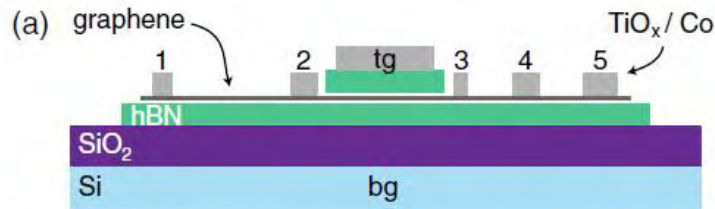
Nature of spin relaxation + spin FET action

Vertical spin transport junctions

Probe proximity to ferromagnetic metals



State of the art: Groningen group

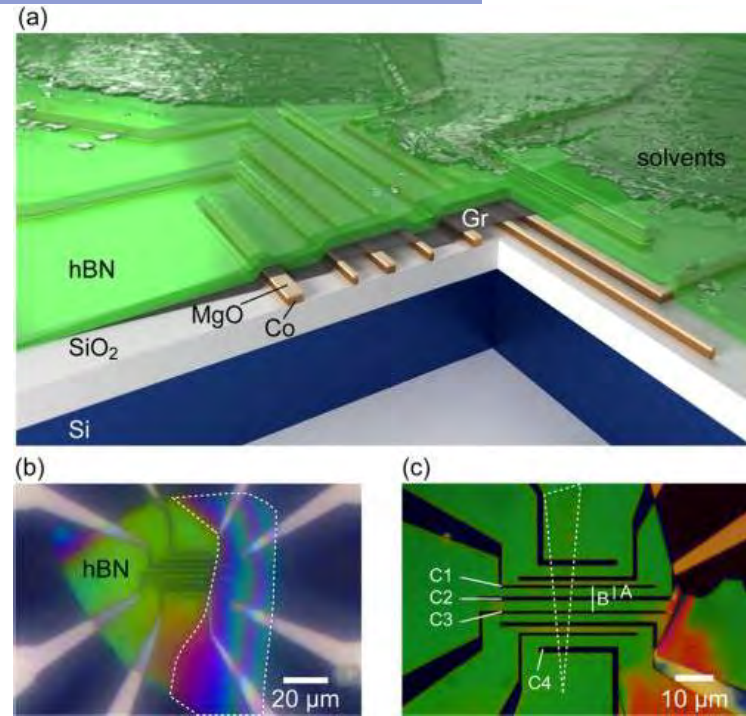
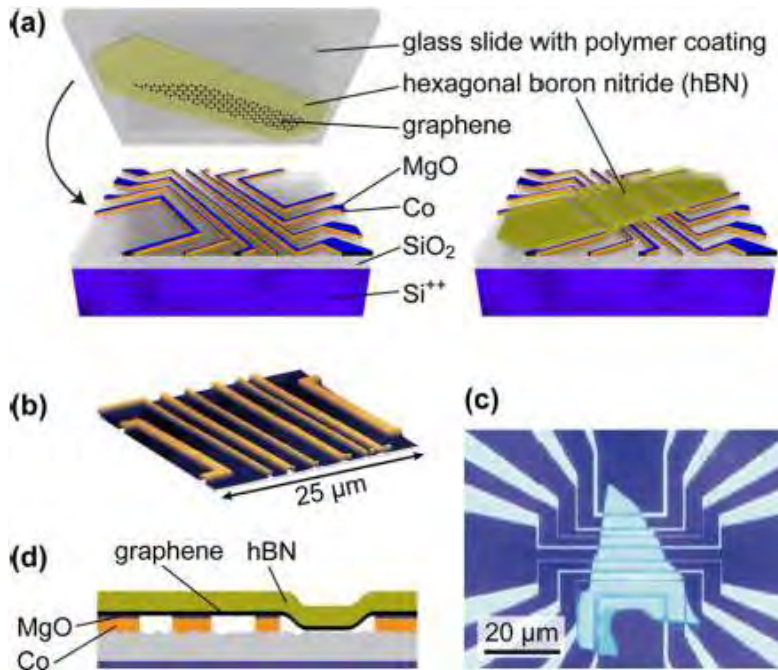


- Partially encapsulated graphene [1]
- $\mu \approx 23000 \text{ cm}^2/\text{Vs}$ at 4.2 K
- Outer regions affect spin transport
- First fully encapsulated w SL h-BN [2]
- $\mu \approx 8600 \text{ cm}^2/\text{Vs}$ at RT
- Low yield (only one made)

[1] M.H.D. Guimarães, P.J. Zomer, J. Ingla-Aynés, J.C. Brant, N. Tombros, and B.J. van Wees, *Phys. Rev. Lett.* **113**, 086602 (2014)

[2] M. Gurram, S. Omar, S. Zihlmann, P. Makk, C. Schönenberger, and B. J. van Wees, *Phys. Rev. B* **93**, 115441 (2016)

State of the art: Aachen group

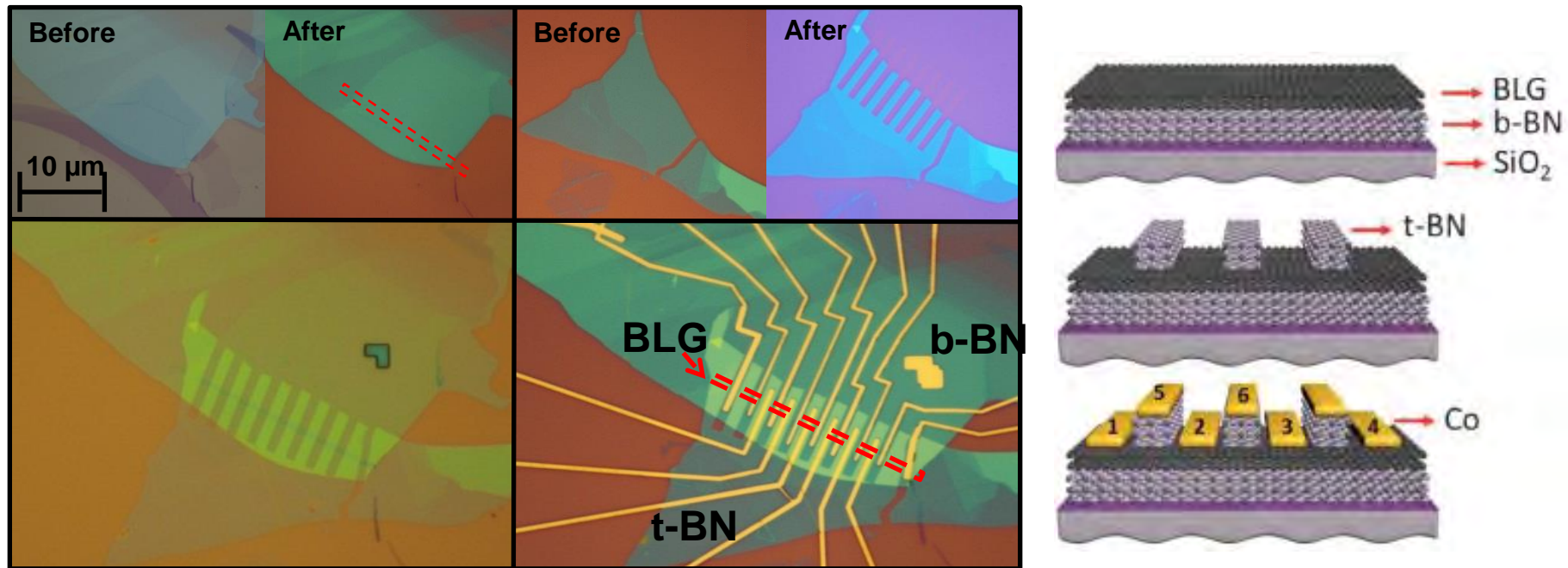


- Bottom -up fabrication technique
- $\mu \approx 20000 \text{ cm}^2/\text{Vs}$ at RT (best quality)
- MgO tunnel barrier exposed after deposition, fixed architecture

*M. Drögeler, F. Volmer, M. Wolter, B. Terrés, K. Watanabe, T. Taniguchi, ... and B. Beschoten, Nano Lett. **14**, 6050 (2014)*
*M. Drögeler, C. Franzen, F. Volmer, T. Pohlmann, L. Banszerus, M. Wolter, ... and B. Beschoten, Nano Lett. **16**, 3533 (2016)*

Novel high-quality architecture

Flexible architecture based on pre-patterned top hBN
Series of fully encapsulated regions + contact regions
Large diffusion lengths ($6\ \mu\text{m}$ @ RT, $10\ \mu\text{m}$ @ 2 K)



A. Avsar, I. J. Vera-Marun, J. Y. Tan, G. K. W. Koon, K. Watanabe, T. Taniguchi, S. Adam, and B. Ozyilmaz, "Electronic Spin Transport in Dual-Gated Bilayer Graphene," *NPG Asia Materials* 8, e274 (2016)

Effect of hBN encapsulation

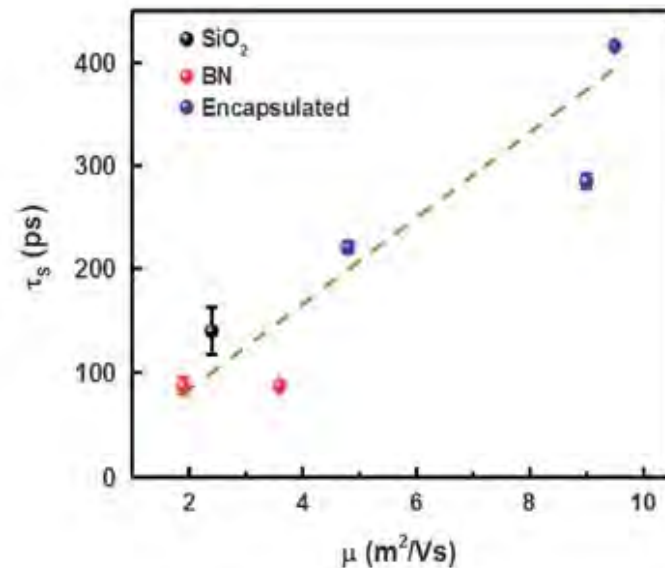
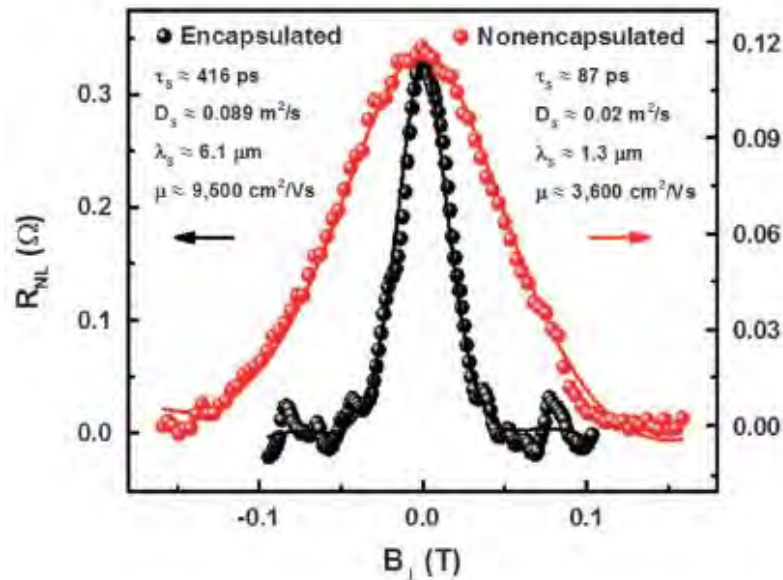
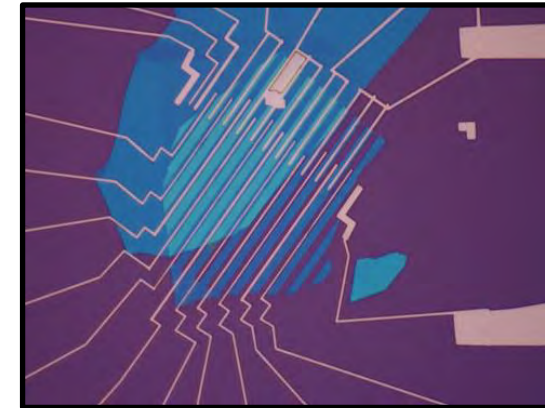
Non-encapsulated: similar to BLG on SiO_2

→ substrate issues such as roughness & charged impurities not the main source of spin relaxation

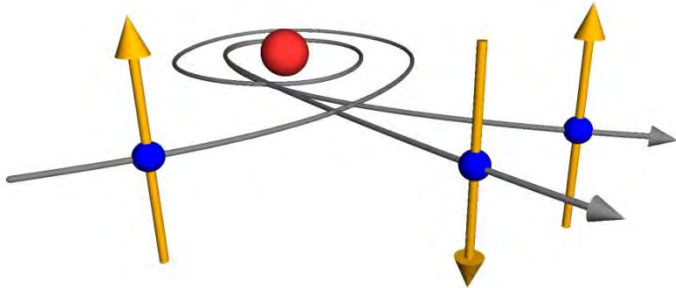
Encapsulated: 5x spin lifetime, 4x spin signal

→ Protection against residues during fabrication

→ Contacts not the main limitation



Recent spin relaxation mechanism: resonant impurity scattering

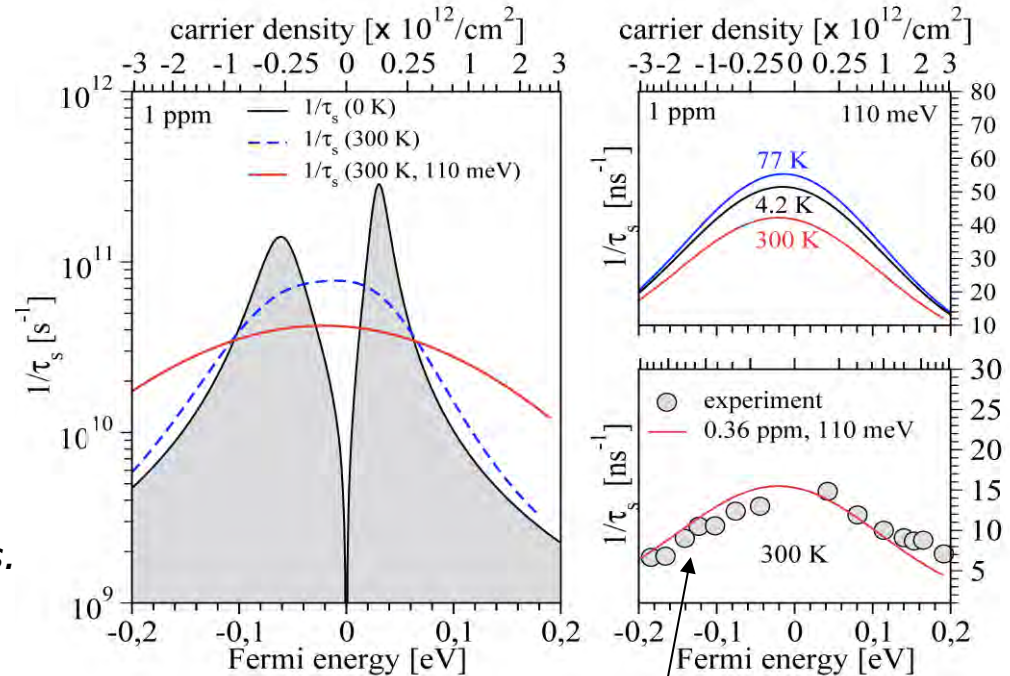


Kochan, D., Gmitra, M. & Fabian, J. *Spin Relaxation Mechanism in Graphene: Resonant Scattering by Magnetic Impurities*. *Phys. Rev. Lett.* **112**, 116602 (2014).

Non-monotonic energy dependence
of spin relaxation time

Magnetic impurities also evidenced
by weak localization experiments

Phys. Rev. Lett. **110**, 156601 (2013)



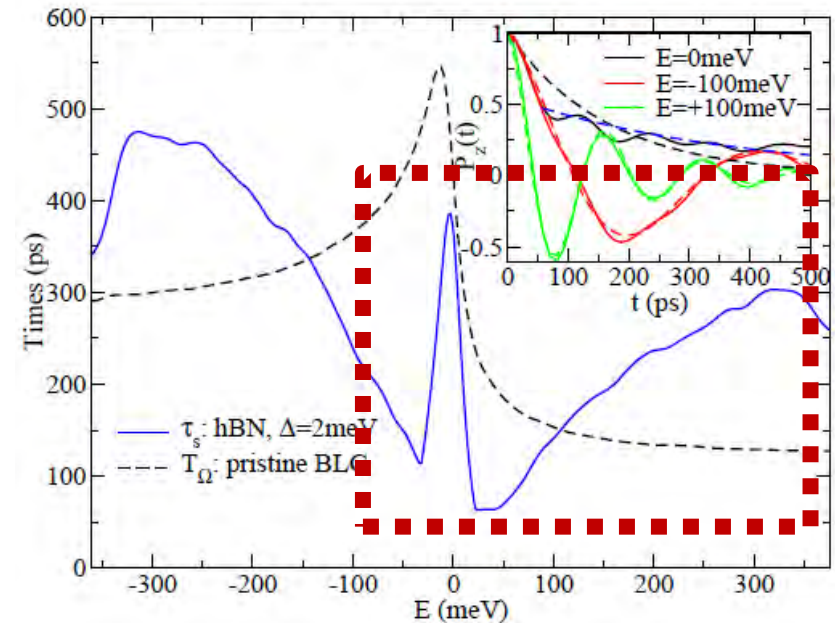
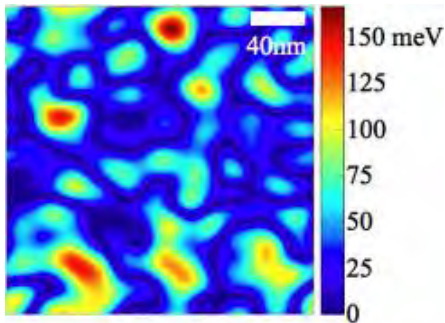
Able to fit SLG experiment from

Wojtaszek, M., Vera-Marun, I.J., Maassen, T. & van Wees, B.J. *Phys. Rev. B* **87**, 81402 (2013)

Recent spin relaxation mechanism: reinforced Dyakonov-Perel

Only Dyakonov-Perel mechanism and
e-h puddles (no magnetic impurities)

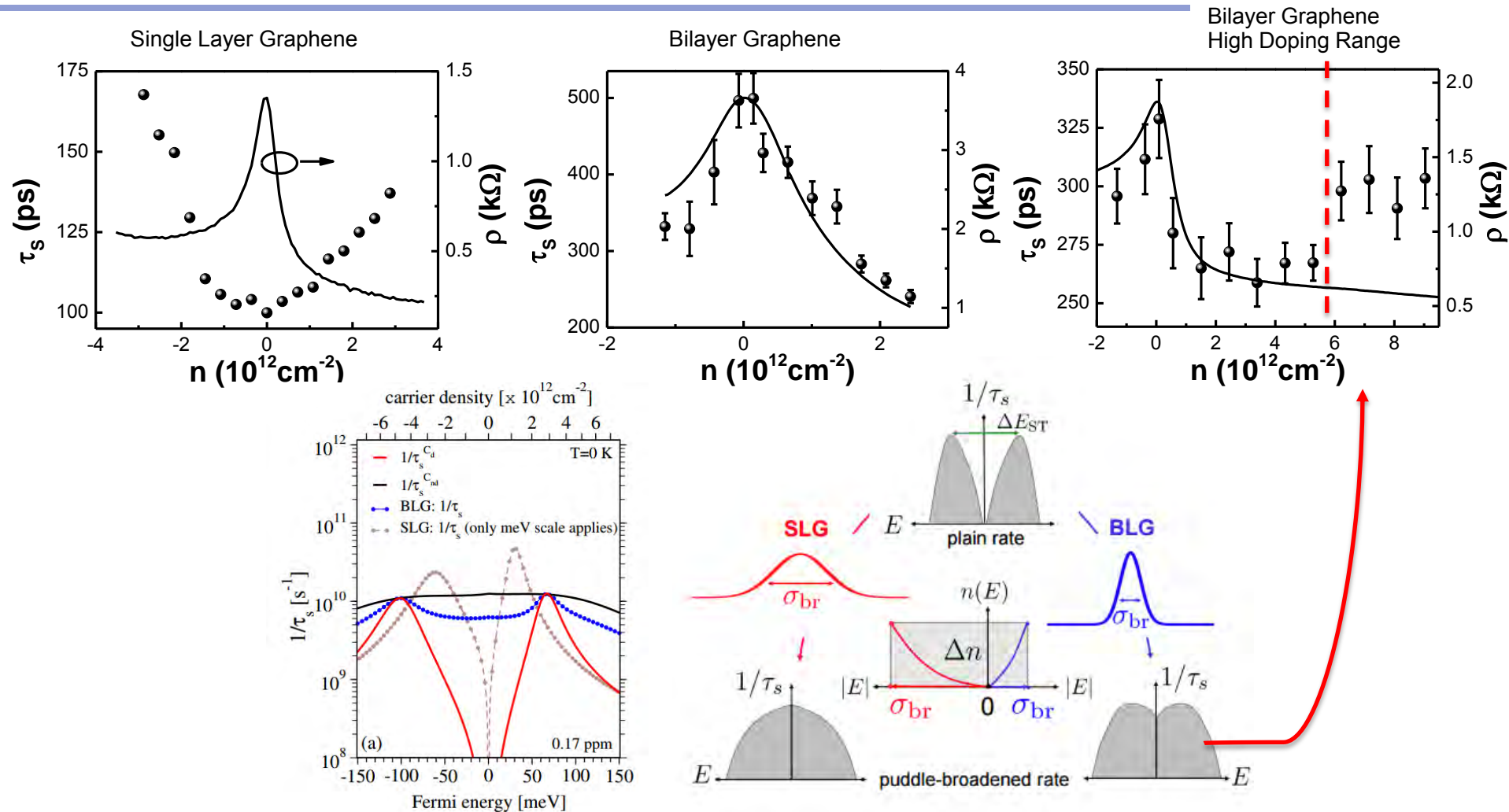
Non-monotonic energy dependence
of τ_s for high-quality bilayer graphene



Van Tuan, D., Adam, S. & Roche, S. Spin dynamics in bilayer graphene: Role of electron-hole puddles and Dyakonov-Perel mechanism. *Phys. Rev. B* 94, 041405(R) (2016)

Van Tuan, D., Ortmann, F., Cummings, A. W., Soriano, D. & Roche, S. Spin dynamics and relaxation in graphene dictated by electron-hole puddles. *Scientific Reports* 6, 21046 (2016)

Spin Transport in BLG



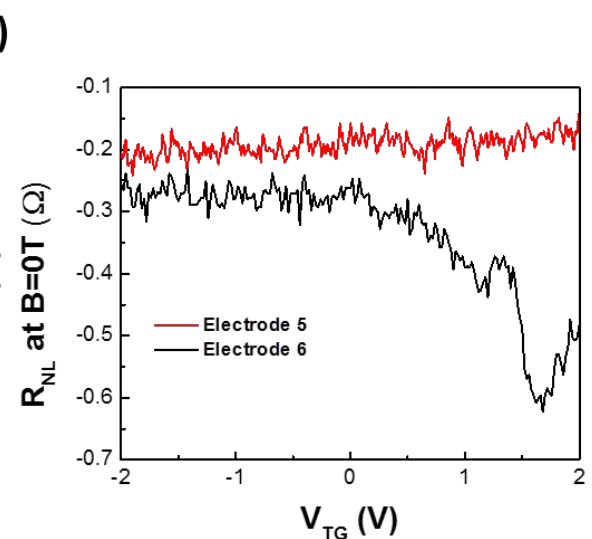
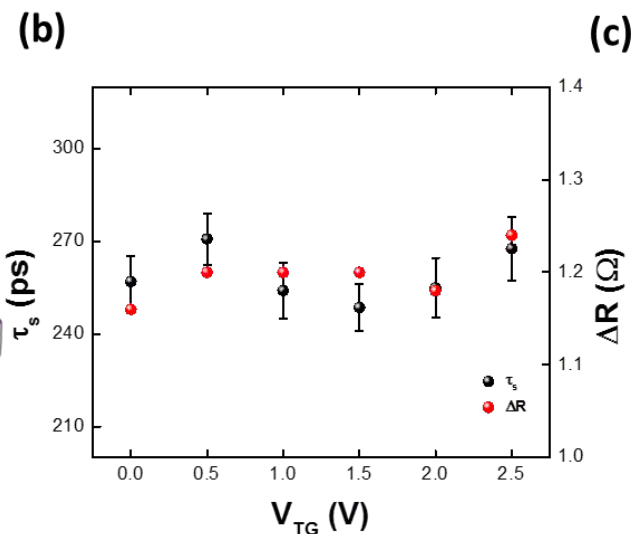
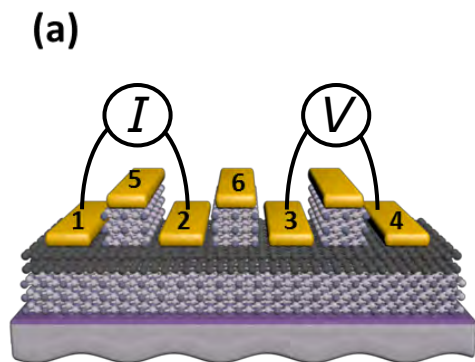
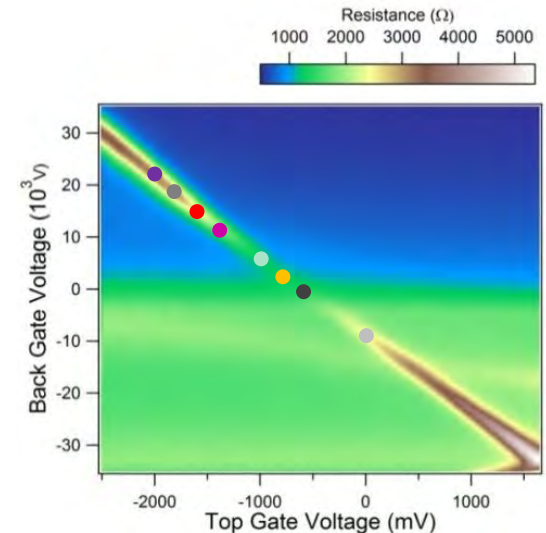
Kochan, D., Irmer, S., Gmitra, M. & Fabian, J. Resonant Scattering by Magnetic Impurities as a Model for Spin Relaxation in Bilayer Graphene. *Phys. Rev. Lett.* **115**, 196601 (2015).

Spin Transport in dual-gated BLG

$\mu \approx 24000 \text{ cm}^2/\text{Vs}$ @ 2K

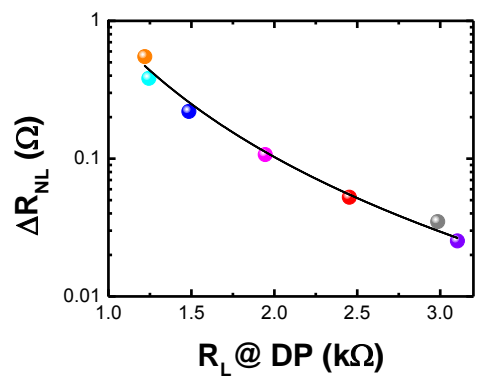
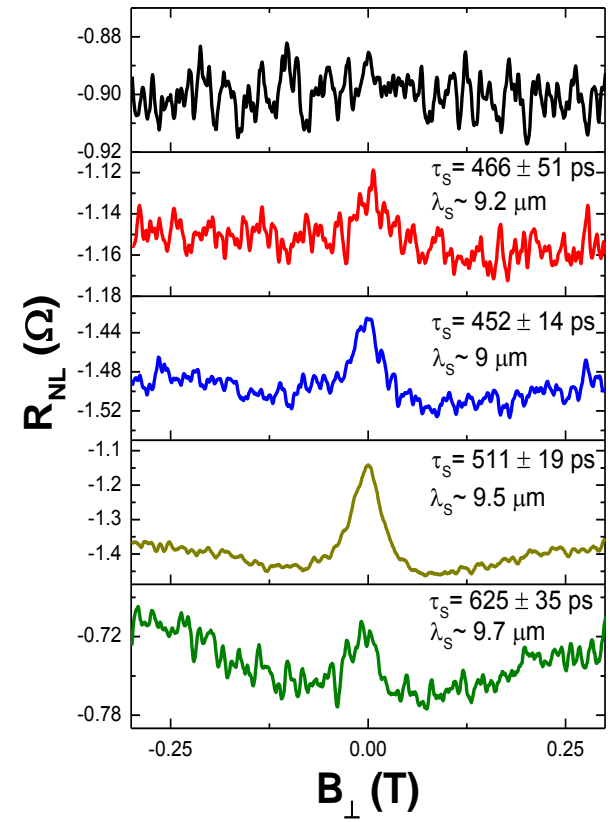
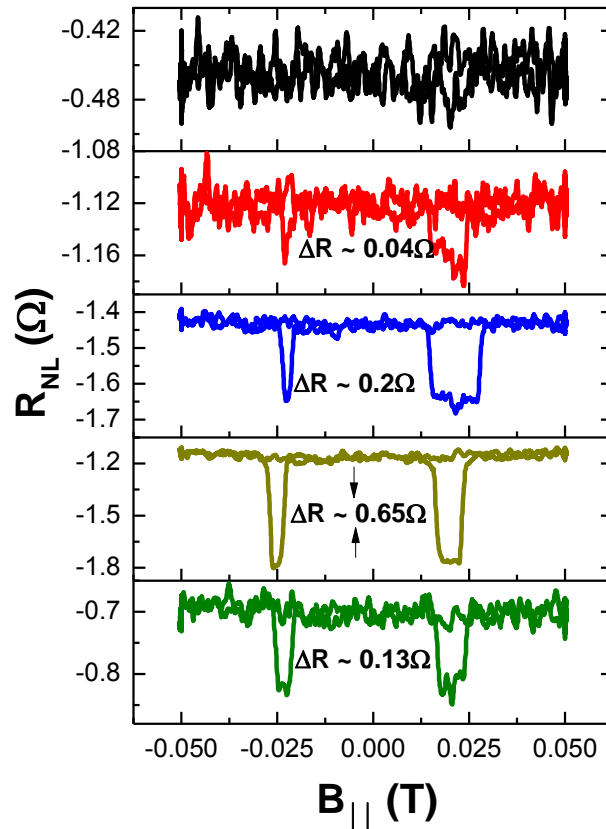
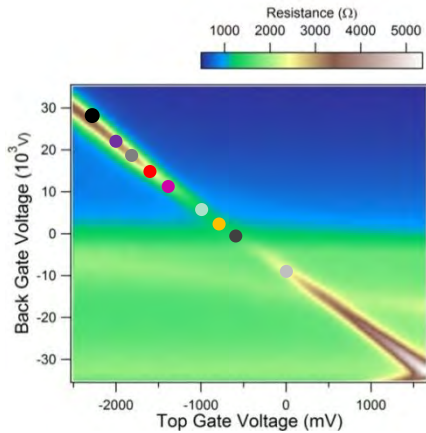
Local control of band-gap (#5) has no affect on nonlocal spin transport

A gap in the nonlocal region (#6) modulates the spin signal \rightarrow spin FET



Spin Transport in dual-gated BLG

- Modulation $>10\times$ spin FET in presence of band-gap @ 2K
- Large $\lambda \sim 10 \mu\text{m}$, $\tau \sim 0.5 \text{ ns}$ \rightarrow no additional spin scattering
- $\mu \approx 24000 \text{ cm}^2/\text{Vs}$ (best quality)



Take-home message (1)

Key role of residues & adsorbates, not substrate or contacts

First observation of non-monotonic energy dependence for BLG spin lifetime, consistent with recent models

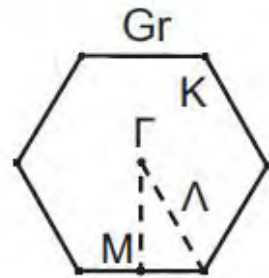
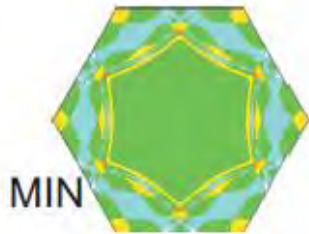
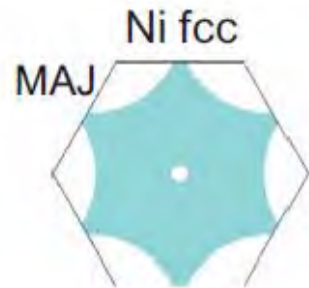
Spin signal modulation $> 10x$ via FET action

A. Avsar, I. J. Vera-Marun, J. Y. Tan, G. K. W. Koon, K. Watanabe, T. Taniguchi, S. Adam, and B. Ozyilmaz, "Electronic Spin Transport in Dual-Gated Bilayer Graphene," NPG Asia Materials 8, e274 (2016)

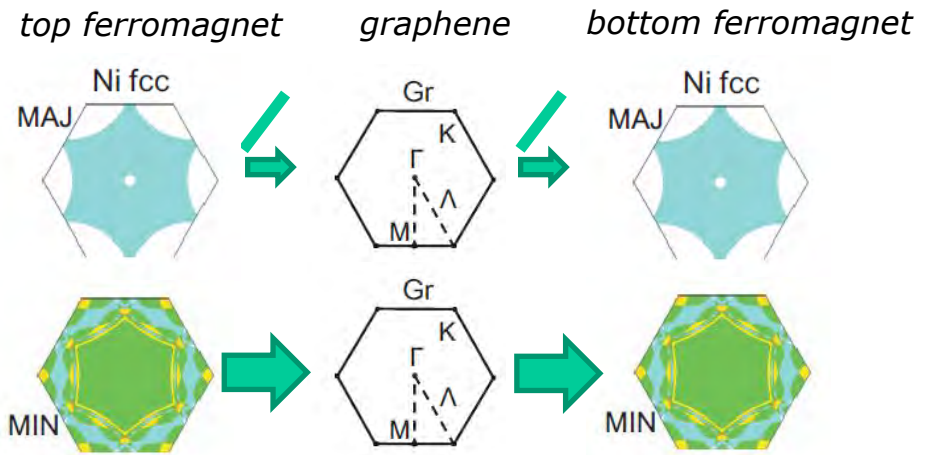
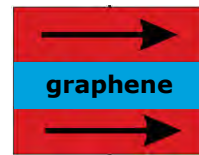


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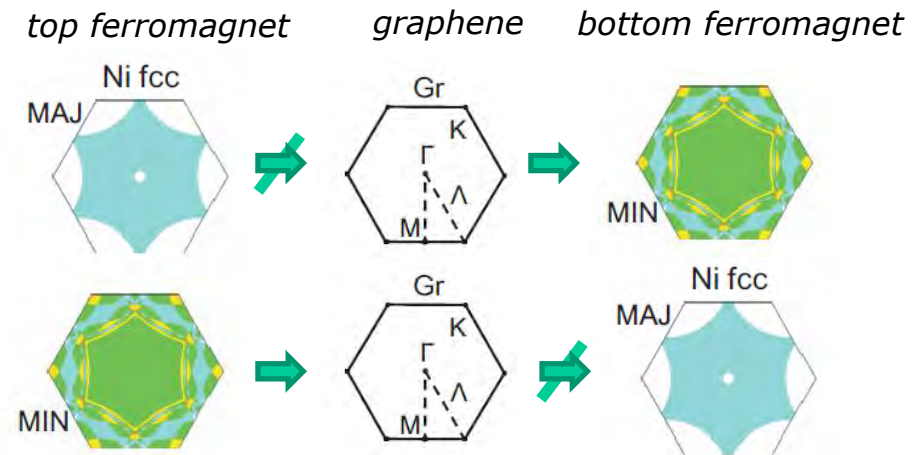
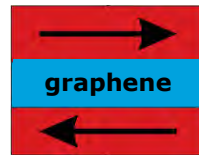
Vertical devices: perfect spin filter?



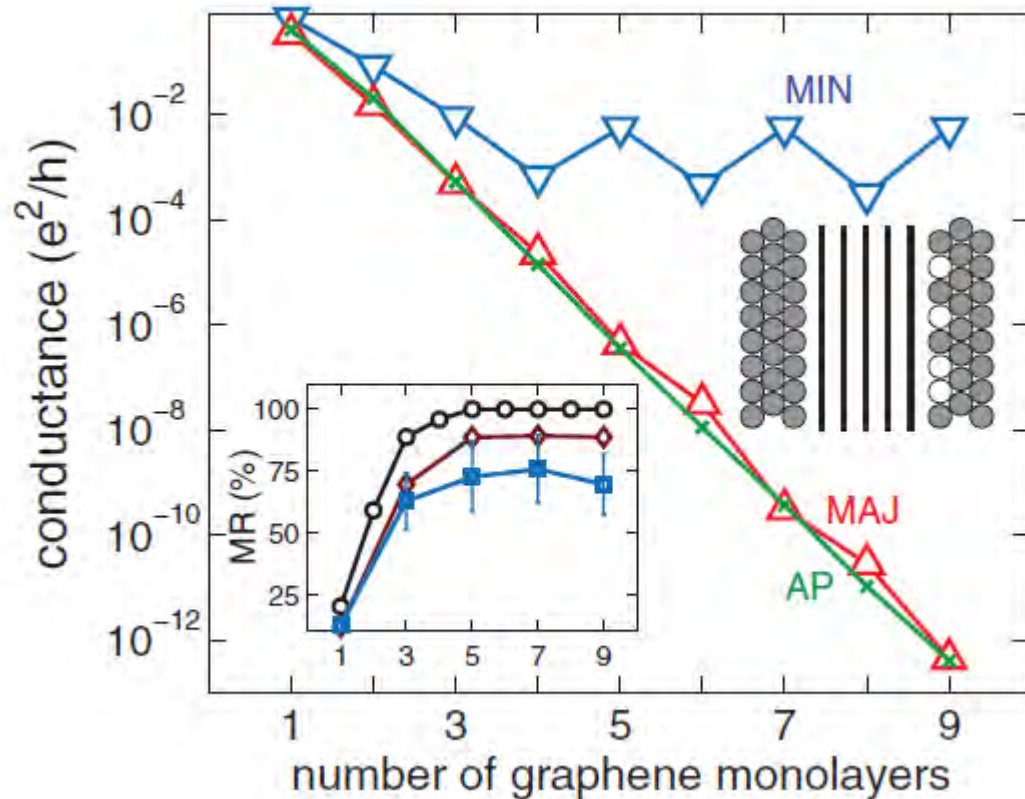
parallel state



anti-parallel state

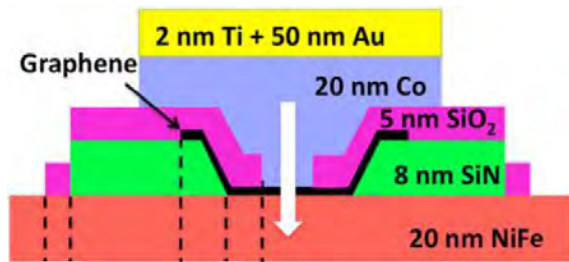


Vertical devices: perfect spin filter?



✓ 100% MR expected for few-layer graphene

Earlier experiments: monolayer spacer

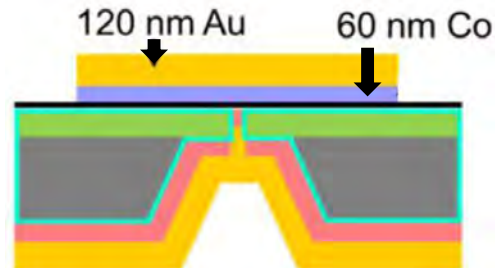


bottom-up fabrication

$\sim 50 \text{ k}\Omega \cdot \mu\text{m}^2$

tunneling, symmetric

MR: 2%

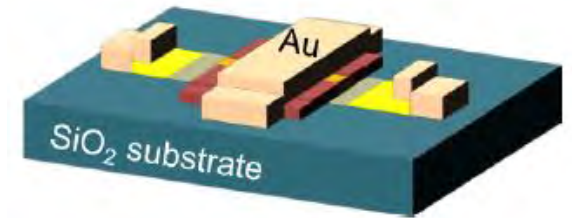


suspended membrane

$\sim 4 \text{ }\Omega \cdot \mu\text{m}^2$

tunneling, asymmetric

MR: 0.8 - 3.4%



'flip-transfer' method

$\sim 0.2 \text{ }\Omega \cdot \mu\text{m}^2$

linear, metallic

MR: 0.3 - 4.6%



magnetoresistance
far below theoretical predictions

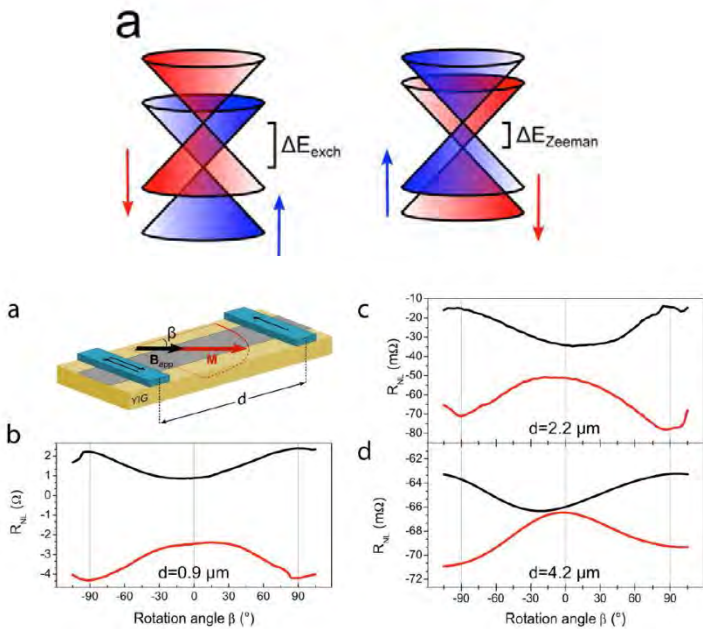
Left to right:

E. Cobas et al. (Naval Res. Labs), *NANO LETT.* 12, 3000-3004 (2012) (possible oxidation of the ferromagnet)

Wan Li et al. (Cornell), *PHYS. REV. B* 89, 184418 (2014) (no oxidation)

Jae-Hyun Park and Hu-Jong Lee, *PHYS. REV. B* 89, 165417 (2014) (no oxidation)

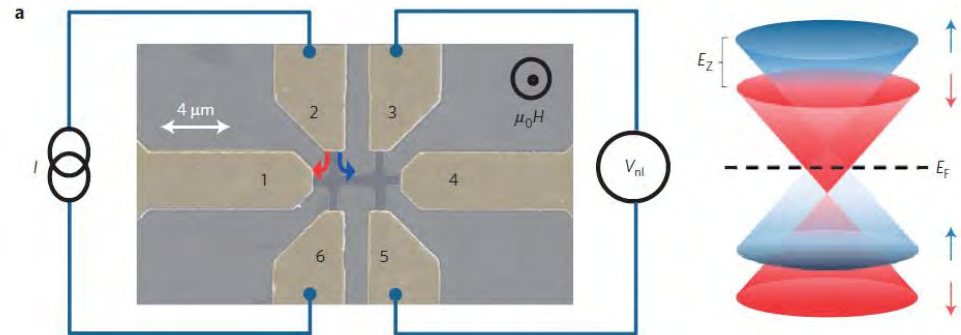
Proximity-induced spin splitting predicted and observed experimentally



$\sim 0.2\text{T}$ B_{exch} for graphene on YIG from lateral spin transport

J.C. Leutenantsmeyer et al, 2D Materials 4, 014001 (2017)

- ✓ expect that the combined effect of doping and proximity-induced spin splitting will determine FM/graphene/FM device behaviour



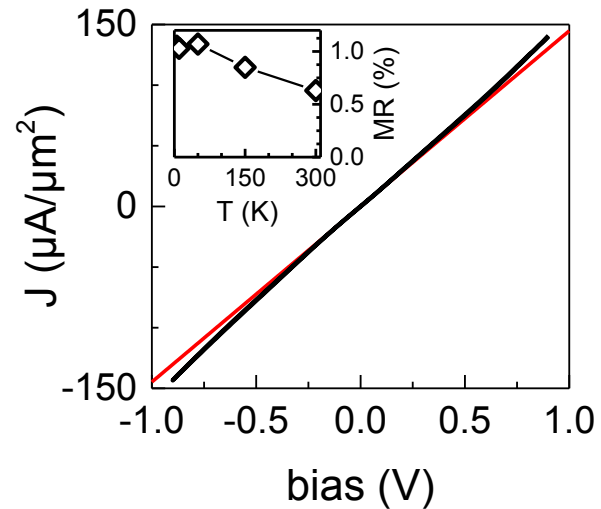
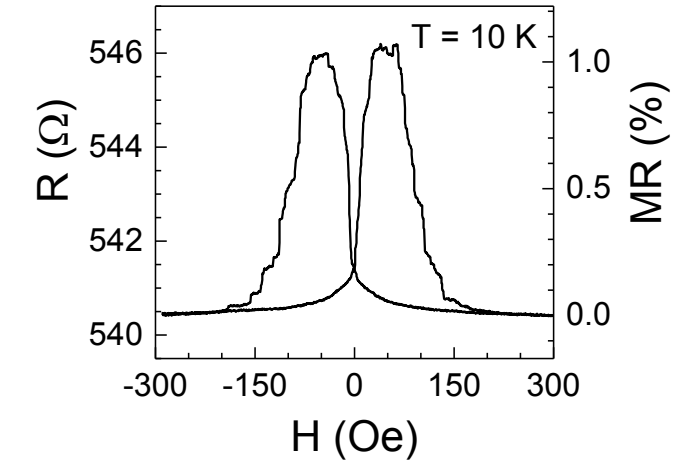
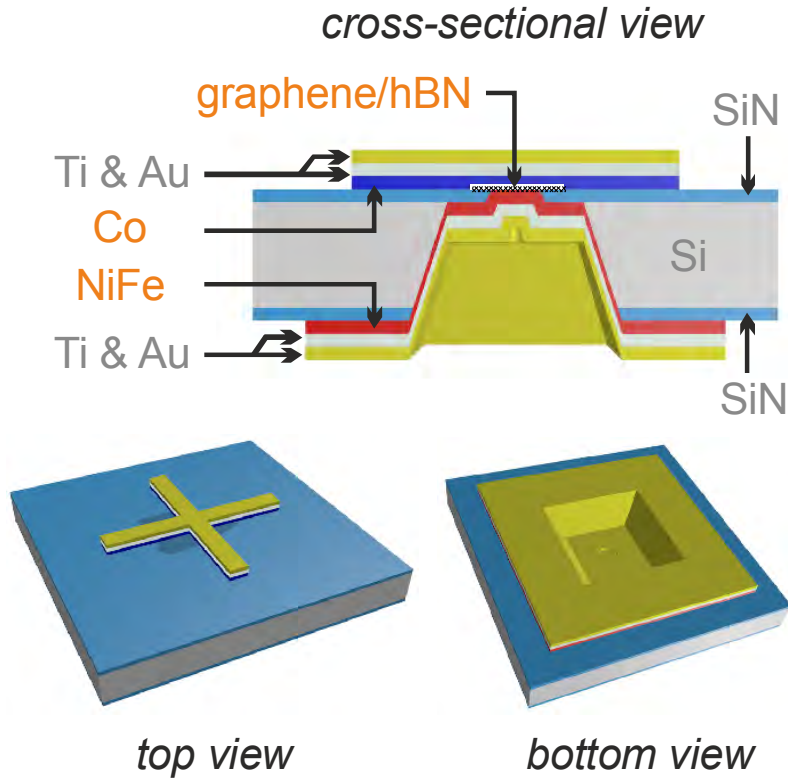
$> 14\text{ T}$ magnetic exchange field in graphene on EuS seen from spin Hall effect

P. Wei et al, *Nature Mater.* DOI: 10.1038/NMAT4603

- ✓ theory predicts $\sim 50\text{-}80\text{ meV}$ spin splitting in graphene in proximity to ferromagnets

Yang, H. X. et al. *Phys. Rev. Lett.* **110**, 046603 (2013).

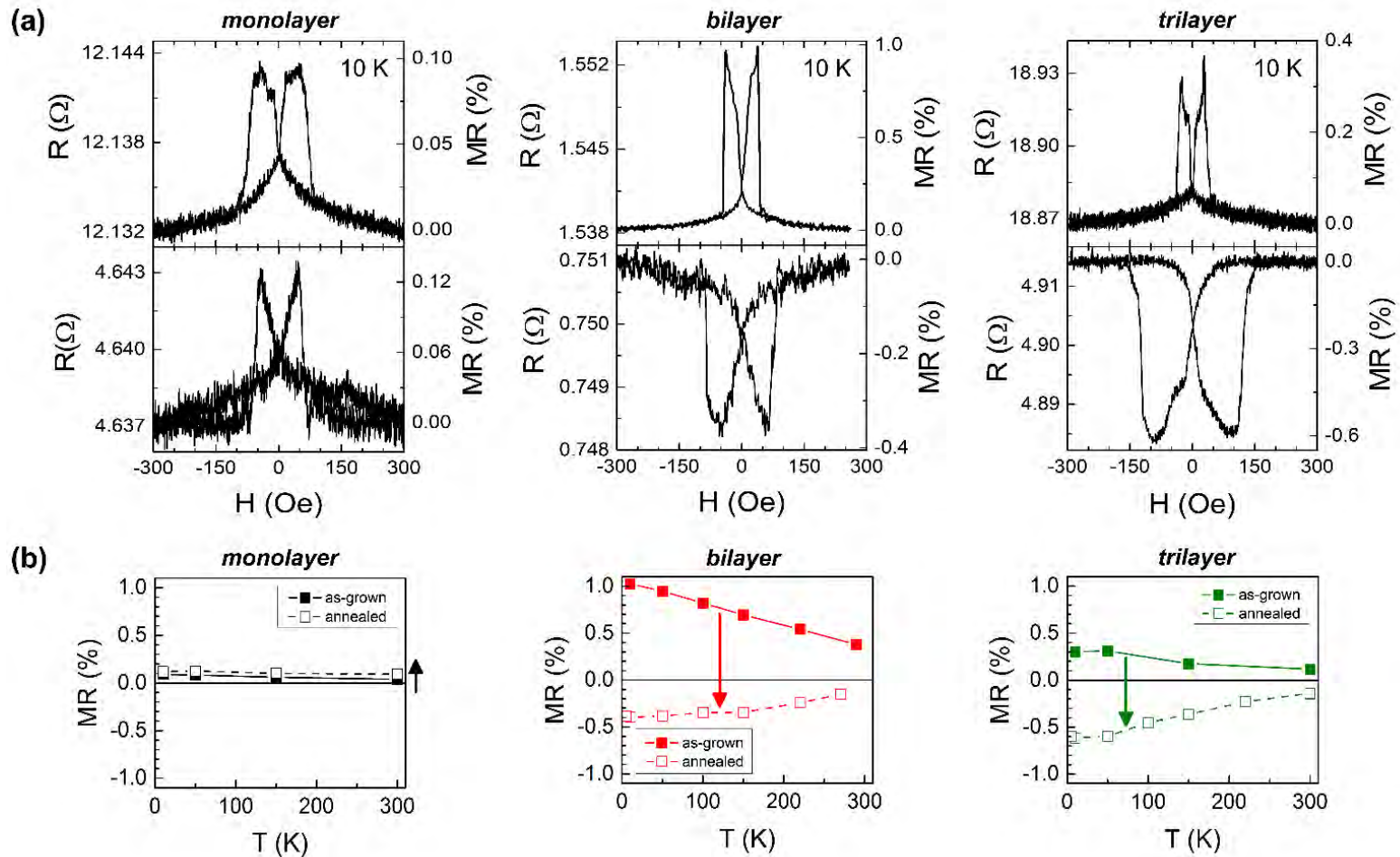
Vertical FM-graphene-FM junctions with 1 to 4 layers of graphene



- ✓ same device architecture, same junction dimensions etc, only change the number of graphene layers

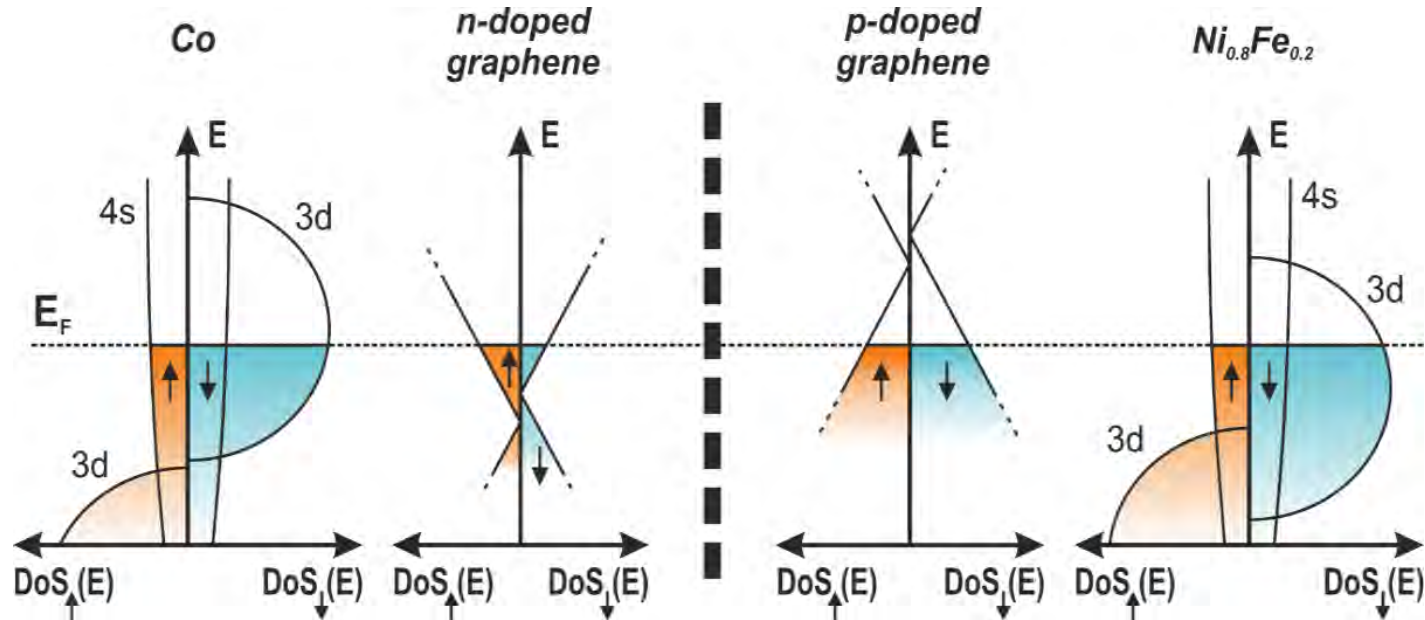
- ✓ tested with BN as a std barrier $\sim 10\%$ P

Different behaviour for mono and bi/tri-layers



- ✓ monolayer graphene: good spacer but small MR, similar to earlier studies;
- ✓ Bi- and trilayer: MR INVERSION after optimizing the FM/G contact

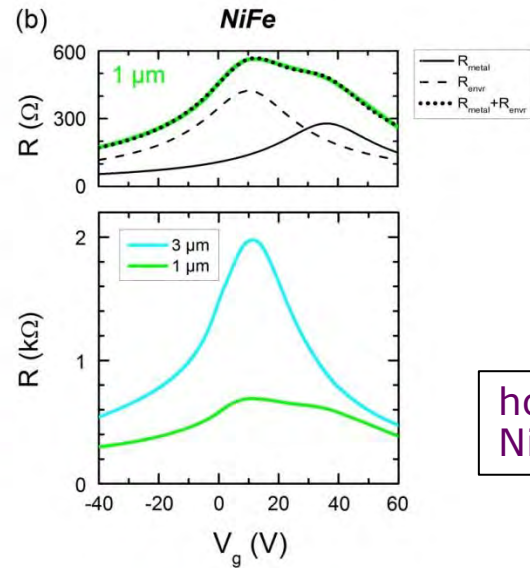
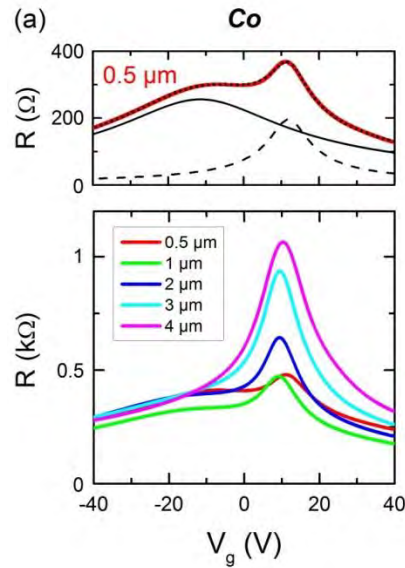
Graphene layers adjacent to different FM metals become different weak ferromagnets



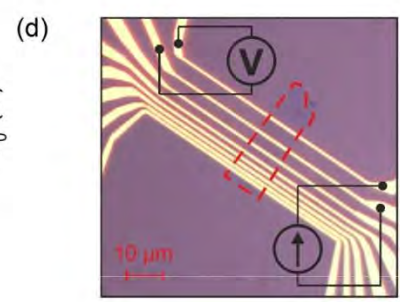
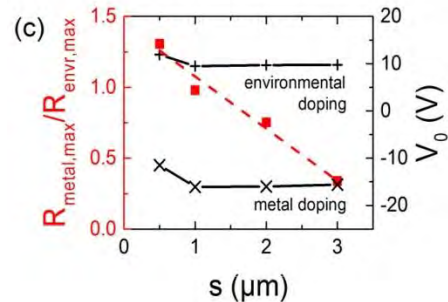
experimental data consistent with the combined effect of doping and proximity-induced spin splitting

What about doping from the metal and proximity-induced spin splitting?

electron doping by Co

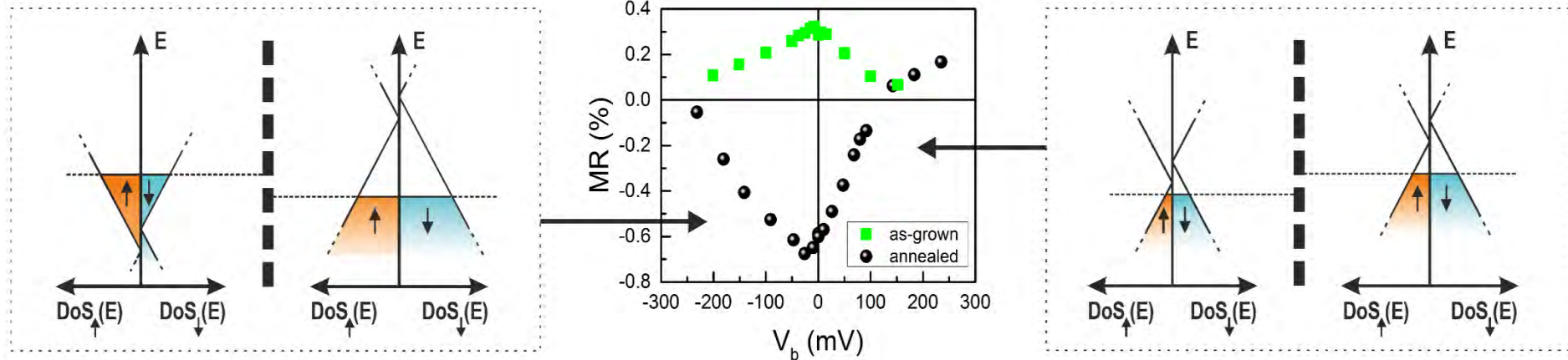


hole doping by NiFe



- ✓ doping of graphene by metal contacts is well known
- ✓ doping by Co and NiFe confirmed in our experiments

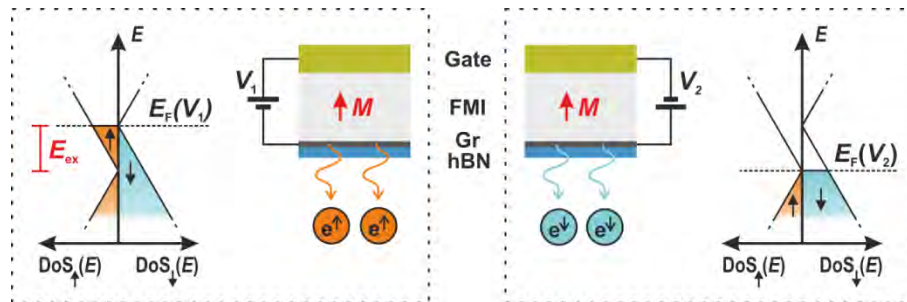
Graphene layers adjacent to different FM metals become different weak ferromagnets



- ✓ spin inversion back to positive MR under small bias, ~ 100 meV
- ✓ corresponds to the Fermi level shift due to doping by Co

Take-home message (2)

- monolayer graphene acts as efficient vertical spacer but does not provide significant MR
- bi- or tri-layer graphene: layers adjacent to FM electrodes become weak ferromagnets determining device behaviour
- *implication:*



Pablo Asshoff
Jose Sambricio
Artem Mishchenko
Andre Geim
Irina Grigorieva

Aidan Rooney
Alexander Rakowski
Sarah Haigh
Sergey Slizovskiy
Ernest Hill
Vladimir Falco

Asshoff, P. *et al.* Magnetoresistance of vertical Co-graphene-NiFe junctions controlled by charge transfer and proximity-induced spin splitting in graphene. *2D Mater.* (2017)



Thank you for your attention

